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Be^7 content in snow

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With 4 figures in the text

SUMMARY

The radioactive isotope Be^7, which is produced in the atmosphere by cosmic radiation, has been extracted from collected snow samples by means of ion exchange. The intensity of the Be^7 radiation has been measured with a scintillation spectrometer. The average concentration of Be^7 atoms in the three snow samples was 4.3×10^3 atoms/cm^3 (volume of water).

Introduction

Since attention has been drawn to the radioactivity that is induced in the atmosphere by the cosmic radiation, Arnold and Al-Salih [1] in Chicago have recently established the existence and measured the amount of Be^7 in rainwater and snow. They found an average concentration of 6×10^3 atoms/cm^3. Since then similar measurements have also been carried out in Bombay by Goel *et al.* [2], who obtained the value of 2.4×10^3 atoms/cm^3.

Both the isotope Be^7 with a half-life of 53 days and the isotope Be^{10}, with a half-life of 2.5×10^6 years, are assumed to be produced, when neutrons of high energy strike oxygen and nitrogen nuclei. Be^7 decays by electron capture and 11 % of the disintegrations lead to the emission of 479 keV gamma quanta (Fig. 1).

As is well known, the intensity of the cosmic radiation varies with latitude in such a way that a broad equatorial zone around the earth shows weaker intensity than the (magnetic) polar caps. The distinction between these three areas of remarkably uniform intensity is rather sharp. Theoretical calculations of the expected concentration of Be^{10} in rain water have been made. Assuming a precipitation of 90 cm of water a year, Goel *et al.* [2] obtained the approximate result of 12×10^3 Be^{10} nuclei/cm^3 at equatorial latitudes and 60×10^3 Be^{10} nuclei/cm^3 at high latitudes.

Since only two intensity measurements of cosmic ray produced Be^7 have been made so far, determination of the Be^7 concentration in other places can be of interest, e.g. for examining the existence of the predicted latitude effect. To these ends a preliminary investigation has been made.

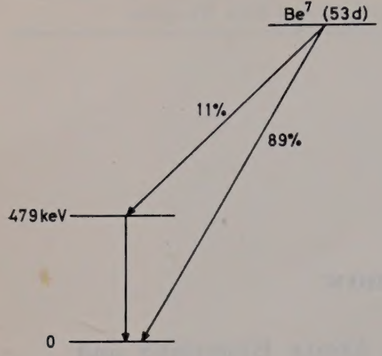

Fig. 1. Decay scheme of Be^7.

Chemical procedure

Three samples have been taken, each consisting of about 75 kg newly fallen snow. The snow was melted and the isolation of the beryllium in the water was performed by means of ion exchange [3]. An arrangement as shown in Fig. 2 was used. The tube was filled with water and a small piece of glass wool placed in the bottom of the column which then was filled to a height of 15 cm with Amberlite IR 100 and covered with a little glass wool.

In order to activate the ion exchanger, about 200 ml of 5 *N* hydrochloric acid was allowed to flow slowly through the column which then was washed with water until the wash-water gave no reaction for chloride ions.

To the melted snow a known amount (10–20 mg) of beryllium in the form of sulphate was added and the pH adjusted to 2.5–3 with hydrochloric acid in order to avoid hydrolysis. This solution was allowed to flow through the ion exchanger at a rate of 50 ml a minute. The beryllium was slowly eluted with 150–200 ml 5 *N* hydrochloric acid and the eluate evaporated to dryness on a water bath.

If beryllium is precipitated as $Be(OH)_2$ with ammonia in the presence of ethylenediamine tetraacetic acid (EDTA) most interfering ions such as iron and aluminium will, according to Brewer [4], remain in solution. The only ions which will interfere are titanium and silicium but these are not likely to be present in more than trace quantities. The isolation of the beryllium was performed in the following manner. The residue from the eluate was dissolved in 10 ml 1 *N* hydrochloric acid. To the solution 5 ml 2 *M* ammoniumchloride and 40 ml 10 per cent disodium ethylenediamine tetraacetate were added. After the addition of 5 drops of 0.05 per cent thymolblue the solution was heated to about 80°C and precipitated with conc. ammonia until the solution turned blue and was filtered through a hardened filterpaper after 12 hours standing.

Since the precipitate will clog to the beaker walls it was necessary to rinse the beaker walls with a few ml of boiling 0.5 *N* hydrochloric acid. To this solution conc. ammonia was added and the mixture was allowed to stand for about ten minutes, after which it was filtered through the same filter as above. The precipitate was washed fifteen times with 2 % cold ammonium chloride solution, which had been made alkaline towards thymol-blue with ammonia, and then once with water. After drying, the filter was ignited in a platinum crucible and weighed as BeO. The recovery calculated on the added amount of beryllium sulphate was 95–98 %.

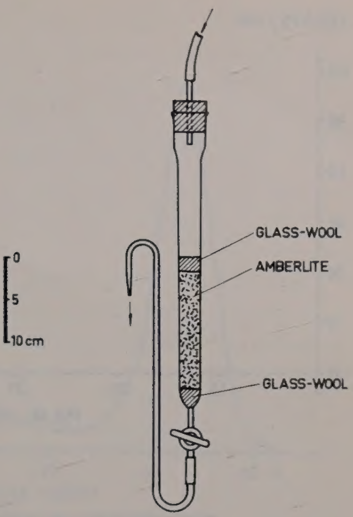

Fig. 2. The ion exchanger used in the chemical procedure.

Measurement of the Be^7 activity

The samples, which took up 3–6 mm of a test tube of 5 mm radius, were examined with a single channel scintillation spectrometer of conventional design. As a detector we used a NaI (Tl) well type crystal of the following dimensions: Diameter $1\frac{3}{4}''$, height $2''$, well diameter $\frac{3}{4}''$, well depth $1\frac{1}{2}''$. The crystal was attached to the photomultiplier and shielded against light in such a way that the protective lid of the counter could be opened and preparations made without any light penetrating into crystal-photomultiplier assembly. The crystal and the photomultiplier were enclosed in a lead envelope. To reduce the recorded background even more, a radiation shield consisting of lead and iron was built around the counter. With this, the background was diminished by about 70 % to 35 counts/min and the accuracy of the measurements were increased to a higher degree.

In order to eliminate as much as possible any drifting with time that might occur and to facilitate the absolute calibration, the intensity of a strong Be^7 source was measured in the spectrometer immediately after the determination of the intensities of the sample and of the background in the appropriate energy interval. The Be^7 source (about $2 \times 4 \times 4$ mm^3) that had been obtained from cyclotron irradiated lithium was used the whole time as a normal, to which the intensities of the several samples were referred.

Fig. 3 shows the photopeak of the spectrum obtained from one of the samples, after the background had been subtracted. The statistical errors are also shown. The curve in the figure is the normalized distribution that was obtained with the strong Be^7 source.

After determining the ratio (q) between the intensity of the Be^7 sample and the intensity of the Be^7 normal for the several plotted points of the photopeak, the weighted mean and the corresponding statistical error were computed.

The result of the measurements is shown in Table 1. Sample A was measured twice. Certain corrections have also been included in Table 1. Some have been applied

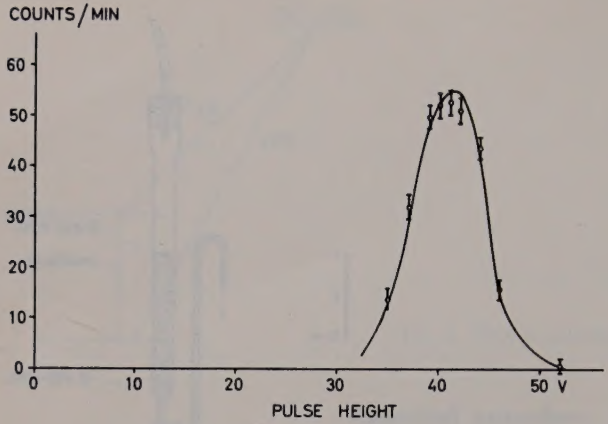

Fig. 3. The photopeak of Be^7.

Table 1.

Sample	$q \times 10^3$	Corr. in %		q
		geom.	chem.	
A A	1.09 ± 0.05 1.00 ± 0.05	0.5	6 ± 2	$(1.12 \pm 0.05) \times 10^{-3}$
B	1.68 ± 0.05	0.8	6 ± 2	$(1.79 \pm 0.06) \times 10^{-3}$
C	2.61 ± 0.04	2.0	6 ± 2	$(2.82 \pm 0.07) \times 10^{-3}$

to eliminate the differences between the vertical extent of the Be^7 normal and the sample. These were computed from measurements that had been carried out with the strong Be^7 normal at several heights above the bottom of the well crystal.

Furthermore the probable yield of the chemical procedure was measured in the following way. The intensity of an artificial, rather strong Be^7 sample was compared with the intensity of the Be^7 normal before and after it had been diluted with 82 kg of water and, like the snow samples, had been allowed to pass through the chemical process. The calculation yielded the result 94 %.

The calibration of the normal

If n pulses from a radioactive isotope are recorded by a scintillation arrangement provided with a differential discriminator, the disintegration rate D can be determined from the following formula:

$$n = D \varrho \omega \varepsilon(E) \eta(E). \tag{1}$$

Here ϱ represents the probability of a certain nuclear transition that leads to the emission of a gamma quantum of energy E, $\varepsilon(E)$ is the detection efficiency for quanta that strike the scintillation crystal, and $\eta(E)$ stands for the fraction of the spectrum that falls within the channel of the discriminator. The effective solid angle that the crystal occupies is represented by ω.

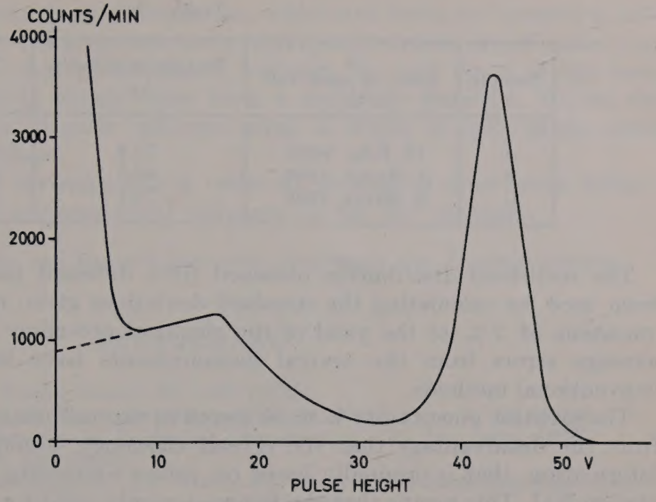

Fig. 4. Spectrum of Be^7, used in the determination of η.

The calibration was carried out with two different scintillation arrangements. In one calculation the measurements were made with the same spectrometer as was the relative measurement. The Be^7 normal was placed 2 mm from the bottom of the well crystal and the whole beryllium spectrum was recorded with a channel width of 1.60 volts (Fig. 4). The value of η was calculated to be 0.408 ± 0.004, after the two areas of the beryllium spectrum had been computed with a planimeter. An 8.30 volt wide channel placed across the photopeak was used in determining n. The total efficiency $\omega\varepsilon$ of the well crystal for 479 keV gamma radiation has been computed by Goel *et al.* [2] and is reported by them to be 41.7 %. For Be^7, $\varrho = 0.11$. The disintegration rate of the Be^7 normal can then be computed. We thus obtain

$$D = (0.877) \pm 0.012) \times 10^6 \text{ decays/min.}$$

The other calibration was made with a spectrometer having an ordinary cylindrical NaI (Tl) crystal with a diameter of $1\frac{1}{2}$ inch and a height of 1 inch. The Be^7 normal was placed 5.0 cm from the crystal and the number of pulses in a 8.55 volt wide channel across the photopeak was recorded. The spectrum was determined with a 1.65 volt channel width; a stronger Be^7 source was used in this measurement. The value of η was determined in the same way as before. The effective solid angle was geometrically computed and the value of the efficiency $\varepsilon = 0.428$ was taken from Bell [5]. The disintegration rate D was then calculated to be

$$D = (0.90 \pm 0.08) \times 10^6 \text{ decays/min.}$$

Results and errors

From the value of the disintegration rate of the Be^7 normal at the time of calibration the amount of Be^7 atoms/cm^3 (water) in the snow samples could be calculated. The Be^7 concentration of the samples was referred to the time of the corresponding snow fall (Table 2).

Table 2.

Sample	Date of snow fall	Weight of sample kg	No. of Be⁷ atoms per cm³
A	19 Feb. 1956	72.3	$(3.05 \pm 0.15) \times 10^3$
B	1 March 1956	80.5	$(3.75 \pm 0.14) \times 10^3$
C	3 March 1956	76.7	$(6.15 \pm 0.18) \times 10^3$

The statistical distribution obtained from different parts of the photopeak has been used for calculating the standard deviations given in the table. In addition a variation of 2 % of the yield of the chemical procedure has been assumed. These average errors from the several measurements have been treated according to conventional methods.

The greatest uncertainty is to be found in the calibration. Both procedures suffer from the disadvantage that the crystal efficiency is computed theoretically from information, that is originally based on values concerning the absorption of gamma rays in NaI. This means that the figures are only valid for an ideal crystal without a scattering and reflecting aluminium envelope. This envelope ought to increase the effective solid angle a little.

Discussion

It is true that the observed average concentration of 4.3×10^3 Be^7 atoms/cm^3 has only been obtained from a small number of measurements, but yet a certain comparison may be made with the corresponding results obtained in Chicago and Bombay.

Concerning the Bombay measurements not only the mean value, 2.4×10^3 atoms/cm^3, but also the maximum value among the 8 samples is less than the mean value found in Uppsala, which is in accordance with the predicted latitude effect.

Chicago (50° N Magn) and Uppsala (54° N Magn) both lie almost on the southern edge of the high latitude plateau of cosmic flux. In Chicago an average concentration of 6×10^3 Be^7 atoms/cm^3 has been found. The values for the 22 samples range from 1.0×10^3 to 16×10^3 atoms per cm^3, indicating large variations between different rains. The Bombay measurements, however, do not show such large variations ($0.9 \times 10^3 - 3.8 \times 10^3$ atoms/cm^3). Arnold and Al-Salih point out that the yields of the chemical procedure, used in Chicago, range from 20 to 80 %.

The three samples collected in Uppsala also show varying intensities. The sample C is considerably more radioactive than A and B. It can therefore be of interest to examine somewhat closer the meteorological conditions prevailing on the different occasions.

The meteorological maps [6] show that the area of precipitation from where sample A came, had moved north, although with low speed, while sample C came from an area of precipitation that had followed a stronger northerly wind. Yet the difference in intensity is not necessarily due to difference in the places of origin since the structures of the precipitations were quite different. In the former case the snow fell in connection with the warm front penetrating towards north. In the latter the precipitation fell on the rear end of a depression where probably the stronger vertical winds cause a more efficient washing down of the radioactive particles, especially through the better mixture between the higher and lower layers of the troposphere.

Sample B came from warm front precipitation, which had fallen in connection with a depression, which had moved at an unusually high speed towards east. C came from the area of precipitation, that subsequently followed the cold front of the same depression, the snow-bearing clouds came from a northerly direction. Hence, the samples B and C represented quite different areas of origin, in spite of the small interval of time between them.

We plan to extend our investigation in order to be able to draw more definite conclusions regarding the meteorological influence on the Be^7 contents.

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